

Interactive comment on "Global carbonyl sulfide (OCS) measured by MIPAS/Envisat during 2002–2012" *by* Norbert Glatthor et al.

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[12pt,a4paper]article [dvips]color Response to reviewer 2:

Many thanks for reading our manuscript and your helpful comments. Please find below our responses describing how the manuscript has been modified with respect to your annotations. Blue passages denote changes or updates in the revised manuscript.

Comment:

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"The very detailed documentation of the spatiotemporal variability of OCS is attractive. But a rigorous statistical treatment of the OCS data, especially in the upper troposphere, is required to justify the results of the current manuscript."

We reply to this below in the context of the major comments.

Major comments:

"1. There have not been any quantitative analysis of the retrieval characteristics. A figure like Figure 3 in Millan et al. [2015, ACP, doi:10.5194/acp-15-2889-2015] is needed to justify the sensitivity of their retrievals to OCS at different altitudes. E.g. How do the sensitivity profiles (i.e. Jacobians) and the averaging kernels look like? What is the a priori concentration? These information have not been presented in Glatthor et al. [2015] nor the current manuscript. However, these information are critical if the authors want to discuss the OCS variability in the upper troposphere or below, as to show how much of the retrieved values actually come from the a priori and the measurement."

Reply: Thank you for pointing this out. We added a plot with MIPAS OCS averaging kernels to Figure 1, which we discuss in the last paragraph of Section 2.2. We do not use optimal estimation but a Tikhonov first order regularization, and the a priori concentration is zero at all altitudes. Our OCS data do not depend on this particular number, because our regularization does not push the result to the a priori but only smooths the retrieval. Any other altitude-constant a priori would lead to the same results, because our regularization constrains only the shape of the profiles but not the values.

"2. The retrieval error analysis is not complete. In Section 2.2, only an estimate of the total errors in the troposphere (50 ppt) and the stratosphere (120 ppt) are presented. The authors explain that the measurement noise is the dominant error. I assume what has been taken as "total errors" in this manuscript is the sum of the "random error" and the forward model parameter errors, defined in von Clarmann et al. [2003]'s Eqs (2) and (3) respectively. von Clarmann et al's Eqs (2) and (3) are the same as Eqs (3.30) and (3.18) of Rodgers [2000] respectively, and do not include the smoothing error (Eq. 3.29 of Rodgers [2000]), which is partly due to the deviation from the a priori and partly due to the Twomey-Tikhonov constraint. Indeed, the smoothing error was not discussed in von Clarmann et al. [2003]. In the current manuscript, the authors used a height-independent constant profile as the a priori. But the OCS concentration varies strongly with height across the tropopause. Therefore, the smoothing error due to the deviation from the a priori should depend on the vertically constant concentration they have assumed. Furthermore, the authors should also mention the error due to ambient temperature."

Reply: In Section 2.2 we indeed give only two estimates of the total error in the troposphere and in the stratosphere. But we refer to Table 1, where the total errors and several error components are given with a better height resolution. We did not want to repeat the content of the whole table in the text. The meaning of the total error has already been given in the captions of Figure 1 and now has also been added to the last but one sentence of Section 2.2. The smoothing error is not contained in our error analysis, because we chose to characterise our retrieval setup by the averaging kernels instead. Even Rodgers does not claim that error estimation is only complete with the smoothing error included. In Rodgers (2000, Sect. 3.2.1) it reads "For the purpose of carrying out an error analysis, the retrieval can be either regarded as an estimate of a state smoothed by the averaging kernel rather than an estimate

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of the true state, or as an estimate of the true state, but with an error contribution due to smoothing". We have chosen the first alternative for reasons discussed in von Clarmann (2014). The error due to the temperature uncertainty is very small and also given in Table 1.

"3. In addition to Comment 2, the 1000 micron band used in this work and Glatthor et al. [2015] for the OCS retrieval is 100 times weaker than the strongest OCS absorption band at 2040 micron that has been used by the IASI and TES teams. The OCS absorption signals at some selected altitudes (e.g. 7 km, 10 km, 20 km, etc) should be compared to the instrument noise, in a similar way in Figure 1 of Millan et al. [2015], to illustrate that the OCS signal is strong enough for retrieval purpose. "

Reply: We assume the reviewer refers to the bands at 860 and 2040 cm⁻¹. Thank you for pointing this out, but please note that both IASI and TES are nadir sounders. For limb emission sounding of the upper troposphere the band at 860 cm⁻¹ is not weaker but actually about 10 times stronger than the strongest OCS absorption band at 2040 cm⁻¹. In addition to the radiances spectral noise has also to be taken into account, which for MIPAS spectra is about 10 times higher in the 860 cm⁻¹ region. Thus, both bands seem to be equally well suited. However, since the spectral signatures in the 2040 cm⁻¹ region become saturated for upper tropospheric observations, we decided to use the band at 860 cm⁻¹. We added a new Figure 1 containing the OCS signatures at 10 and 20 km along with spectral noise to the manuscript, which illustrates that the band is strong enough for retrieval purpose. This figure is discussed in paragraph 2 of Section 2.2.

"4. The fact that the seasonal patterns are obtained with a constant a priori is quite promising but the authors should also plot the evolution of the error terms in the same way as in their Figure 5 to show that the seasonal patterns are not results of errors."

Reply: Due to the heavy computational load required, a full error analysis was performed for selected MIPAS scans only. The only error estimate available for every MIPAS scan is the estimated standard deviation (ESD). Thus, we plotted the evolution of the ESD of the bin-averaged OCS values in the same way as in Figure 5. These plots generally show lower variations and no correlation with the seasonal pattern of the volume mixing ratios (see attached Figure).

"5. The comparison between MIPAS OCS and other OCS measurements are not consistent. The SPIRALE data have been convolved with the averaging kernel before comparing MIPAS whereas ACE-FTS and MkIV have not. The authors explain that it was because the vertical resolution of SPIRALE is higher. However, in additional to the degradation of the vertical resolution, the effect of the a priori in the MIPAS OCS is also applied to the SPIRALE through the averaging kernel. Therefore, the averaging kernel is applied either to all datasets or to none."

Reply: The constraint applied to MIPAS measurements by the IMK processor is a first order Tikhonov constraint. Contrary to usual optimal estimation retrievals, where the a priori covariance matrix is used as a regularization matrix, the absolute OCS amount is in the null space of the regularization matrix. Thus, the constraint can only reduce the altitude resolution but cannot push the retrieval towards the a priori. However, since MIPAS Aks become vertically displaced in the stratosphere, they have also been applied to the MkIV and ACE-FTS profiles, and the discussion in Sections 3.1 and 3.3 has been slightly adjusted.

Minor comments:

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"1. Page 1, Line 17: Should "tropospheric OCS" be actually "upper tropospheric OCS"? The authors mostly discuss the OCS in the upper troposphere near 10 km or 250 hPa. But in the abstract (and in the text), the authors sometimes refer to "tropospheric OCS" (e.g. for the trends). The authors should clarify whether they are actually referring the upper tropospheric OCS or really tropospheric OCS, say, in the model simulations or inferred from HCN or ozone data."

Reply: We changed the wording into "upper tropospheric OCS" on page 1, lines 17 and 20.

"2. Page 2, Line 23-24: "A comprehensive compilation of these budget estimations is given in Kremser et al. (2016)." Do you mean Kremser et al. (2015)?"

Reply: No, we mean the review paper of Kremser et al. (2016), as given in the reference list.

"3. Page 3, Line 4-5: How would Lejeune et al. [2011] discuss the trend in 2012? Please check."

Reply: The correct year should have been "2010". However, very recently an updated analysis of the Jungfraujoch trends until the year 2015 has been presented in Lejeune et al. (2017). We now cite the trends given in this publication.

Comment: "4. Page 3, Line 24: Somehow the authors should also mention IASI and TES OCS products for completeness because this manuscript discusses the tropospheric OCS."

Reply: We agree. To account for IASI and TES OCS products, the sentence "Spaceborne OCS measurements of the NASA Aura Tropospheric Emission Spectropmeter (TES) and of the Infrared Atmospheric Sounding Interferometer (IASI) have been presented by Kuai et al. (2014) and by Vincent and Dudhia (2016), respectively." has been added at the end of paragraph 5 of the Introduction.

"5. Page 5, Line 3: What's values of a priori used by retrieval? What did you use for constraint of a priori? Could you show the profile of a priori with uncertainties you used in Figure 1?"

Reply: We used a zero a priori profile and added this information "(zero at all altitudes)" to the manuscript. As already indicated in the preceding sentence of the manuscript, a first order Tikhonov smoothing constraint was applied. The effect of the a priori and the constraint is fully characteriszed by the averaging kernels. We do not think it is necessary to show the zero a priori profile applied.

"6. Page 5, Line 15: 41–48 pptv and 10–26% cannot be both right."

Reply: These values are both right, because the corresponding retrieved OCS mixing ratios are 414.0 pptv (15 km) and 182.5 pptv (20 km).

"7. Figure 2: Is the same a priori profile used at all locations? If not, it may be better to show the a priori profile in each panel."

Reply: The same height-constant a priori profile (zero at all altitudes) is used for each

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MIPAS geolocation.

"8. Page 8, Line 3: Has there been any explanation why there was an increase of OCS concentration at 14 km after 2006?"

As outlined in Section 4.1 of the manuscript, the main increase of OCS at 14 km occurs in the second half of 2005 and in our opinion is due to the change of the observation mode. As correctly observed by the referee, there is a slight additional increase in 2006. We changed the wording in the updated manuscript into "Another slight increase occurs in 2006, just as at 18 km altitude. The reason for this increase, either geophysical or instrumental, is unclear."

"9. Page 11, Line 25: The term "slower convection" is contradictory. Should it be "vertical mixing" or "upwelling"?"

Reply: We changed "slower convection" into "weaker vertical mixing".

"10. Page 13, Line 29-31, "In the northern hemisphere there is a band of enhanced values extending from the tropical Atlantic to the Chinese coast, which reflects the upper end of the Asian Monsoon Anticyclone including westward outflow." Could this high OCS extending from Atlantic to China also result from Arabic anticyclone?"

Reply: As far as we know, there is no "Arabic anticyclone" in contrast to the "Asian monsoon anticyclone". Instead, there are two modes of the Asian monsoon anticyclone, the "Iranian mode" and the "Tibetan mode". During one monsoon season the anticyclone moves once or several times between the modes, occasionally also

splitting up, followed by westward or eastward outflow. Thus, we think the feature in Fig. 12, which is averaged over the summer months of 9 years, reflects the Asian Monsoon Anticyclone.

"11. The discussion of Asian Monsoon anticyclone (AMA) signature of OCS distribution at UTLS is illuminating on the underlying transport. The authors may want to explain more clearly what mechanisms caused the pattern of enhanced OCS on the north end and low OCS on the top of AMA."

Reply: We do not quite understand, what the referee means here. Is it about differences between 150 and 80 hPa? In this case our interpretation of the plots is that at 150 hPa the AMA extends over a wide subtropical area and that at 80 hPa just the central part of the AMA is still visible.

References:

von Clarmann, T.: Smoothing error pitfalls, Atmos. Meas. Tech., 7, 3023-3034, doi:10.5194/amt-7-3023-2014, 2014.

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